

IN THE UNITED STATES DESIGNATED/ELECTED OFFICE

In re Application of:

Holger von STENZEL et al.

Group Art Unit 1796

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Title: ION-CONDUCTIVE THERMOPLASTIC COMPOSITIONS FOR ELECTROCHROMIC GLAZING

DECLARATION UNDER 37 C.F.R. §1.132

Commissioner for Patents
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Sir:

I, ALEXANDER KRAFT being duly warned, declare that:

I am a citizen of GERMANY, residing in EICHWALDE, Germany.

I am an inventor of the above-captioned application and am, therefore, familiar with the invention described therein and with the grounds for rejection made against the claims of the application in the Office Action mailed 27 December 2010, from the U.S. Patent and Trademark Office.

The following experiments were conducted by me or under my supervision.

1. Comparative examples: ion-conductive PVB foil and electrochromic element produced therefrom according to the state of the art

Comparative Example I: (See Example 1 in the specification on page 9)

An ion-conductive PVB foil was prepared with the following composition:

- 65 % by weight of PVB with a polyvinyl butyral content of 77.5 % by weight, a PVOH content of 20.5 % and a polyvinyl acetate content of 2 % and
- 35 % by weight of the plasticiser tetraethylene glycol dimethyl ether containing lithium trifluoromethane sulphonate as support electrolyte (7.33 % by weight) as well as the UV absorber Tinuvin 571 (0.15 mass %) was extruded on a twin screw extruder with co-rotating screws (manufacturer: Leistritz, type LSM 30.34) equipped with a melting pump and slit die at a temperature of the mixture of 160 °C.

Using this foil, an electrochromic element was produced. For this purpose, a K-glass pane (FTO-coated float glass) was coated electrochemically with tungsten trioxide and a second K-glass pane with Prussian blue. These two panes equipped with the above-mentioned electrochromic films were laminated together with the ion-conductive PVB foil described above (previously conditioned at 23 °C and 50 % relative

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atmospheric humidity) according to the state of the art using a standard autoclave process for composite safety glass. The active switchable surface of this element amounted to 7.5 cm x 18.5 cm (213.75 cm²). After lamination, contacting and sealing of the electrochromic element with an epoxy resin took place. The ion conductivity of the foil in the finished element was determined from the impedance at 40 kHz. A value of $3.3 \cdot 10^{-6}$ S/cm was obtained. During switching of the electrochromic element a strong decrease of the switching capacity occurred.

Comparative example II:

The above procedure was repeated except an ion-conductive PVB foil was prepared with the following composition:

- 65 % by weight of PVB with a polyvinyl butyral content of 65.8 % by weight, a PVOH content of 32.2 % and a polyvinyl acetate content of 2 % and
- 35 % by weight of the plasticiser TGEDME containing lithium trifluoromethane sulphonate as support electrolyte (7.33 % by weight)

The ion conductivity of the foil in the finished element was determined from the impedance at 40 kHz. A value of $3.37 \cdot 10^{-6}$ S/cm was obtained. During switching of the electrochromic element delamination occurred.

2. Practical examples: ion-conductive PVB foil according to the invention

Practical Example 1: (See Example II in the specification on page 10)

An ion-conductive PVB foil with the composition:

- 65 % by weight of PVB with a PVOH content of 20.2 %, a polyvinyl acetate content of 1.8 %, a content of acetal of glyoxylic acid of 0.5 % and a polyvinyl butyral content of 77.5 % and
- 35 % by weight of the plasticiser tetraethylene glycol dimethyl ether containing lithium trifluoromethane sulphonate as support electrolyte (7.33 % by weight) and the UV absorber Tinuvin 571 (0.15 % by weight) was produced as described in comparative example 1.

Using this foil, an electrochromic element was produced as described in comparative example 1. The active switchable surface of this element was 9 cm x 30 cm (270 cm²). After lamination, contacting and sealing of the electrochromic element, as described in comparative example 1, took place. The ion conductivity of the foil in the finished element was determined from the impedance at 40 kHz. A value of $6.9 \cdot 10^{-6}$ S/cm was determined.

The incorporation of the glyoxylic acid group into the polymer chain of the PVB consequently leads to a more than 100% increase in the ion conductivity with an otherwise identical foil composition.

During switching of the electrochromic element no decrease of the switching capacity and no delamination occurred.

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Practical Example 2:

The Example above was repeated except an ion-conductive PVB foil was prepared with the following composition:

- 65 % by weight of PVB with a PVOH content of 18 %, a polyvinyl acetate content of 2 %, a content of acetal of glyoxylic acid of 2 % and a polyvinyl butyral content of 78 % and
- 35 % by weight of the plasticiser tetraethylene glycol dimethyl ether containing lithium trifluoromethane sulphonate as support electrolyte (7.33 % by weight)

The ion conductivity of the foil in the finished element was determined from the impedance at 40 kHz. A value of $6.9 \cdot 10^{-6}$ S/cm was determined.

The incorporation of the glyoxylic acid group into the polymer chain of the PVB consequently leads to a more than 100% increase in the ion conductivity with an otherwise identical foil composition.

During switching of the electrochromic element no decrease of the switching capacity and no delamination occurred.

Practical Example 3:

The above procedure was repeated except an ion-conductive PVB foil was prepared with the following composition:

- 65 % by weight of PVB with a PVOH content of 23.5 %, a polyvinyl acetate content of 2 %, a content of acetal of glyoxylic acid of 1% and a polyvinyl butyral content of 74.3 % and
- 35 % by weight of the plasticiser tetraethylene glycol dimethyl ether containing lithium trifluoromethane sulphonate as support electrolyte (7.33 % by weight)

The ion conductivity of the foil in the finished element was determined from the impedance at 40 kHz. A value of $6.8 \cdot 10^{-6}$ S/cm was determined.

The incorporation of the glyoxylic acid group into the polymer chain of the PVB consequently leads to a more than 100% increase in the ion conductivity with an otherwise identical foil composition.

During switching of the electrochromic element no decrease of the switching capacity and no delamination occurred.

Practical Example 4:

The above procedure was repeated with an ion-conductive PVB foil with the composition:

- 68 % by weight of PVB with a PVOH content of 20.1 %, a polyvinyl acetate content of 2 %, a content of acetal of glyoxylic acid of 0.2 % and a polyvinyl butyral content of 77.7 % and
- 32 % by weight of the plasticiser tetraethylene glycol dimethyl ether containing lithium trifluoromethane sulphonate as support electrolyte (7.33 % by weight)

The ion conductivity of the foil in the finished element was determined from the impedance at 40 kHz. A value of $5.0 \cdot 10^{-6}$ S/cm was determined.

The incorporation of the glyoxylic acid group into the polymer chain of the PVB consequently leads to a more than 50% increase in the ion conductivity with an almost identical foil composition.

During switching of the electrochromic element no decrease of the switching capacity and no delamination occurred.

Practical Example 5:

The above procedure was repeated with an ion-conductive PVB foil with the composition:

- 65 % by weight of PVB with a PVOH content of 20.1 %, a polyvinyl acetate content of 2 %, a content of acetal of glyoxylic acid of 0.2 % and a polyvinyl butyral content of 77.7 % and
- 30 % by weight of the plasticiser tetraethylene glycol dimethyl ether containing lithium trifluoromethane sulphonate as support electrolyte (7.33 % by weight)
- 5 % by weight of the plasticiser triethylene glycol diheptanoate

The ion conductivity of the foil in the finished element was determined from the impedance at 40 kHz. A value of $4.8 \cdot 10^{-6}$ S/cm was determined.

The incorporation of the glyoxylic acid group into the polymer chain of the PVB consequently leads to about a 40% increase in the ion conductivity with an almost identical composition.

During switching of the electrochromic element no decrease of the switching capacity and no delamination occurred.

Practical Example 6:

The above procedure was repeated with an ion-conduction PVB foil with the composition:

- 65 % by weight of PVB with a PVOH content of 20.1 %, a polyvinyl acetate content of 2 %, a content of acetal of glyoxylic acid of 0.2 % and a polyvinyl butyral content of 77.7 % and
- 35 % by weight of the plasticiser tetraethylene glycol dimethyl ether containing lithium trifluoromethane sulphonate as support electrolyte (7.33 % by weight)

The ion conductivity of the foil in the finished element was determined from the impedance at 40 kHz. A value of $6.9 \cdot 10^{-6}$ S/cm was determined.

The incorporation of the glyoxylic acid group into the polymer chain of the PVB consequently leads to a more than 100% increase in the ion conductivity with an otherwise identical foil composition.

During switching of the electrochromic element no decrease of the switching capacity and no delamination occurred.

Practical Example 7:

The above procedure was repeated with an ion-conduction PVB foil with the following composition:

- 70 % by weight of PVB with a PVOH content of 20.1 %, a polyvinyl acetate content of 2 %, a content of acetal of glyoxylic acid of 0.2 % and a polyvinyl butyral content of 77.7 % and
- 30 % by weight of the plasticiser tetraethylene glycol dimethyl ether containing lithium trifluoromethane sulphonate as support electrolyte (10 % by weight)

The ion conductivity of the foil in the finished element was determined from the impedance at 40 kHz. A value of $6.75 \cdot 10^{-6}$ S/cm was determined.

The incorporation of the glyoxylic acid group into the polymer chain of the PVB consequently leads to an 100% increase in the ion conductivity with an otherwise identical foil composition.

During switching of the electrochromic element no decrease of the switching capacity and no delamination occurred.

Tables 1 and 2 (below) summarize the results of the above comparative tests of PVB according to the state of the Art and PVB according to the present invention. The prior art Examples of Table I are closer to the invention than the prior art cited in the Office Action (i.e. Heckner et al. and Papenfuchs et al.)

Table 1. Preparation PVB according to the State of the Art

Content of PVB polymer	subunit 1 of PVB polymer: polyvinyl butyral	subunit 2 of polymer: polyvinyl acetate	subunit 3 of polymer: polyvinyl alcohol (PVOH)	subunit 4 of polymer: polyvinyl acetal of glyoxylic acid	Content of plasticiser	Content of support electrolyte	Ion Conductivity	Stability of electrochromic element (most important parameter)	Remarks
Examples with PVB according to the state of the Art:									
65%	77.5%	2%	20.5%	-	35% TEGDME*	7.33% Li-Triflate**	$3.3 \cdot 10^{-6}$ S/cm	Low: strong decrease of switching capacity	Comparative Example I: See, Example 1 on page 9 of specification: "Standard PVB" (state of the art)
65%	65.6%	2%	32.2% (PVOH around 20% is normal, increased PVOH generally leads to ac samples with fast delamination)	-	35% TEGDME	7.33% Li-Triflate	$3.37 \cdot 10^{-6}$ S/cm	Very low: Delamination	Comparative Example II

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Table 2.0 Preparation of PVB according to the invention

Content of PVB polymer	subunit 1 of PVB polymer: polyvinyl butyral	subunit 2 of polymer: polyvinyl acetate	subunit 3 of polymer: polyvinyl alcohol (PVOH)	subunit 4 of polymer: polyvinyl acetal of glyoxylic acid	Content of plasticiser	Content of support electrolyte	Ion Conductivity	Stability of electrochromic element (most important parameter)	Remarks
Examples with PVB according to the invention:									
65%	77.5%	1.8%	20.2%	0.5%	35% TEGDME	7.33% Li-Triflate	$6.9 \cdot 10^{-6}$ S/cm	High: low decrease of switching capacity	Practical Example 1 (See Example 2 on page 10 of the specification)
65%	78%	2%	18%	2%	35% TEGDME	7.33% Li-Triflate	$6.9 \cdot 10^{-6}$ S/cm	High: low decrease of switching capacity	Practical Example 2
65%	74.3%	2%	23.5%	1%	35% TEGDME	7.33% Li-Triflate	$6.8 \cdot 10^{-6}$ S/cm	High: low decrease of switching capacity	Practical Example 3
68%	77.7%	2%	20.1%	0.2%	32% TEGDME	7.33% Li-Triflate	$5.0 \cdot 10^{-6}$ S/cm	Very high: very low decrease of switching capacity	Practical Example 4
65%	77.7%	2%	20.1%	0.2%	30% TEGDME 5% 3G7***	7.33% Li-Triflate	$4.8 \cdot 10^{-6}$ S/cm	Very high: very low decrease of switching capacity	Practical Example 5
65%	77.7%	2%	20.1%	0.2%	35% TEGDME	7.33% Li-Triflate	$6.9 \cdot 10^{-6}$ S/cm	Very high: very low decrease of switching capacity	Practical Example 6
70%	77.7%	2%	20.1%	0.2%	30% TEGDME	10% Li-Triflate	$6.75 \cdot 10^{-6}$ S/cm	High: low decrease of switching capacity	Practical Example 7

* TEGDME Tetraethylene glycol dimethylether

** Li-Triflate Lithium trifluoromethane sulphonate

*** 3G7 Triethylene glycol diheptanoate

The test results demonstrate that the PVB compositions according to claim 1 result in a significant and unexpected 30-100 % increase in ion conductivity (i.e., from 4.8 to $6.9 \cdot 10^{-6}$ S/cm) compared to the prior art compositions (i.e., 3.3 to $3.7 \cdot 10^{-6}$ S/cm). Moreover, the compositions according to the present invention have a highly stable electrochromic element resulting in a low decrease of switching capacity. In contrast, the prior art compositions have a low stability electrochromic element resulting in either a strong decrease of switching capacity or delamination.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

A handwritten signature in dark ink, consisting of a series of loops and a long horizontal stroke at the end, positioned above a solid horizontal line.

Date: June 22nd, 2011